

# MODELING OF THERMODYNAMIC BEHAVIOR OF THERMOPLASTIC COMPOSITES DURING PREHEATING IN AN INFRA-RED OVEN

E. Kobler<sup>1,2\*</sup>, J. Birtha<sup>1</sup>, B. Cäsar<sup>2</sup>, L. Gahleitner<sup>3</sup>, G. Mayr<sup>3</sup>, K. Straka<sup>2</sup>, C. Marschik<sup>1</sup>, G. Steinbichler<sup>2</sup>, P. Zwicklhuber<sup>4</sup> and S. Schlecht<sup>5</sup>
<sup>1</sup> Competence Center CHASE GmbH, Altenberger Strasse 69, 4040 Linz, Austria,

eva.kobler@chasecenter.at, https://www.chasecenter.at

<sup>2</sup> Institute of Polymer Injection Molding and Process Automation, Johannes Kepler University, Altenberger Strasse 69, 4040 Linz, Austria, <u>bernhard.caesar@jku.at</u>, <u>https://www.jku.at/institut-fuer-polymer-injection-moulding-and-process-automation/</u>

 <sup>3</sup> Research Group of Thermography and NDT, University of Applied Sciences Upper Austria, Stelzhamerstrasse 23, 4600 Wels, Austria, <u>lukas.gahleitner@fh-wels.at</u>, <u>www.thermo-ndt.at</u>
<sup>4</sup> ENGEL Austria GmbH, Steyrer Strasse 20, 4300 St. Valentin, Austria, <u>https://www.engelglobal.at</u>
<sup>5</sup> Covestro Deutschland AG, Kaiser-Wilhelm-Allee 60, 51373 Leverkusen, <u>https://www.covestro.com</u>

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## ABSTRACT

The aim of this work is to present a simulation method to predict the core temperature of a thermoplastic composite material during heating in an infra-red oven. During the experiments, the surface temperature of the composite part is recorded with pyrometers that control the heating process. Therefore, the presented approach considers only the heat transfer from the component surface to the core. For this reason, only one material parameter, the thermal diffusivity, is needed.

Active thermography is used to determine the thermal diffusivity of samples with different degrees of deconsolidation produced during heating tests in an infra-red oven, in order to obtain a general relationship between the thermal diffusivity and the deconsolidation and consequently the temperature.

The values obtained are used to simulate the heating tests performed with the open-source simulation toolbox OpenFOAM®.

## **1 INTRODUCTION**

The interest in thermoplastic composites as lightweight materials has increased in recent years, due to their many advantages over thermoset composites. Some of the most important advantages are (i) high damage tolerance in terms of high fracture toughness, (ii) high impact and fatigue resistance, and (iii) outstanding corrosion and solvent resistance [1, 2].

One advantage related to the processing is the possibility of simultaneously forming and functionalizing a thermoplastic composite part in an injection molding machine. This requires preheating the semi-finished part to a temperature at which the material is formable, i.e. above the glass transition temperature for amorphous polymers and above the melting temperature for semi-crystalline polymers. Due to their efficiency, typically infra-red heaters are used to heat thermoplastic composites prior to forming [3,4]. In this case a consolidated layup consisting of 18 layers of unidirectional (UD) fiber reinforced tape of polycarbonate with 44% carbon fiber by volume is used as semi-finished part.

Simulations allow fast insights into the process and assist its optimization. The complexity of infrared heating, however, can lead to high computational costs if all phenomena, like various forms of heat transfer, reflection scattering of radiation and elusive and non-reproducible effects are modeled [5]. To avoid this, the computational fluid dynamics (CFD) software tool OpenFOAM® is used, where a simple model is implemented by default, that only considers the heat transfer from the component surface to the core of the component.

## 2 MODELING

Fig. 1 shows (one half of) the used tape stack, which is consolidated after stacking and subsequently heated and formed.



 $[0^{\circ}|90^{\circ}|0^{\circ}|90^{\circ}|0^{\circ}|90^{\circ}|0^{\circ}|90^{\circ}|0^{\circ}]_{S}$ 

Figure 1: Consolidated stack of UD Tapes.

To model the heat transfer from the surface of the composite to the core, the Laplace equation is solved:

$$\frac{\partial T}{\partial t} - \nabla \cdot (a \nabla T) = S,\tag{1}$$

where T is the temperature, t is the time, and a is the thermal diffusivity of a mixture of matrix, fibers and - if deconsolidation occurs - air inclusions. The thermal diffusivity is further defined as:

$$a = \frac{\lambda}{c_n * \rho},\tag{2}$$

where  $\lambda$  is the thermal conductivity,  $c_p$  ist the specific heat capacity at constant pressure, and  $\rho$  is the density.

In Eq. (1) *S* describes an optional source term, which is ignored here. In the original model the thermal diffusivity is considered constant [7]. This assumption may be invalid in real heating processes, taking into account that the composite material can deconsolidate when its temperature is increased, causing the formation of air inclusions. Therefore, a function was implemented to consider the dependency of thermal diffusivity on the temperature. The surface temperature of the composite part which was recorded by pyrometers during the experiments was set as boundary condition for the surface of the part. This allowed us to ignore elusive phenomena, like radiation scattering or convection within the surrounding air, and limited the considered simulation domain to the composite part.

## **3 MATERIALS AND METHODS**

#### 3.1 Material

A cross-ply layup of 18 UD tape layers ( $[0^{\circ}|90^{\circ}|0^{\circ}|90^{\circ}|0^{\circ}|90^{\circ}|0^{\circ}|90^{\circ}|0^{\circ}]s$ ) of polycarbonate with 44% carbon fiber by volume (provided by Covestro Deutschland AG, Leverkusen, Germany) was used. To measure the temperature of the stack during IR heating, three thermocouples (Type K) were located within the layup. The sensor tips and wires were consolidated in a previous processing step between layer 1 and 2, 9 and 10, and 17 and 18, respectively. Attempts were made to place the sensors as centrally as possible, with a minimum distance of 2 cm from each other to avoid excessive thickness changes caused by the wires. The total size of the parts was 230 x 150 mm<sup>2</sup> (LxW). For each temperature setting three samples were used.

## 3.2 Heating experiments

To validate the model, experiments were performed with an infra-red oven (IRO BG3, ENGEL Austria GmbH, St. Valentin, Austria) at the LIT Factory in Linz, Austria. In the oven the semi-finished part is heated from both sides. During these experiments the data of both, the thermocouples and the pyrometers measuring the surface temperature during heating process were recorded.

The heating temperatures were set to 200°C, 250°C and 300°C oven temperature. The experiments

were considered to be completed when the measured temperature at each layer was equal to the set temperature  $(\pm 10\%)$  for a period of at least one minute. Three samples were processed for each of the set temperatures.

#### 3.3 Measurement of the thermal diffusivity

The thermal diffusivity of the composite material, which is the only material property required for the simulation (see Eq. (2)), was measured by means of active thermography at the University of Applied Sciences Upper Austria in Wels, Austria. To this end, the thermal diffusion time was experimentally evaluated, based on the linear diffusivity fitting (LDF) approach [6]. The sample was placed within a fixation frame between a thermography camera (quantum detector, FLIR X8400sc) and a flash lamp (PB-G6000, power = 6kJ, heating time = 2 ms), as shown in Fig. 2.



Figure 2: Measurement setup for active thermography to determine the thermal diffusivity of a consolidated tape stack.

In this method, the rise in temperature, caused by the energy input of the flash, on the back side, which is recorded by the IR-Camera, is linearized. By plotting  $\log(T(L, t) - T_0 + \frac{1}{2}\log(t))$  versus  $\binom{1}{t}$  the slope  $\beta$  can be determined, which is defined as [6]:

$$\beta = -\frac{t_d}{4} = -\frac{L^2}{4a},\tag{3}$$

where  $T_0$  is the temperature at the beginning of the measurement,  $t_d$  is the diffusion time, L is the part thickness. Therefore, using equation (3), it is possible to determine the thermal diffusivity indirectly by linearizing the measured temperature rise.

In this work, the method was used to average the thermal diffusivity of the entire part to obtain a representative value. Samples of various degrees of deconsolidation, which were produced at different heating temperatures in the infra-red oven, were investigated to determine a general relationship between thermal diffusivity and deconsolidation, and consequently temperature.

#### **4 RESULTS**

# 4.1 Experimental results

## 4.1.1 Heating experiments

The following graphs show the temperature measured by the thermocouples inside the layup.



Figure 3: Temperature measured by thermocouples inside the layup at 200°C heating.



Figure 4: Temperature measured by thermocouples inside the layup at 250°C heating.



Figure 5: Temperature measured by thermocouples inside the layup at 300°C heating.

Fig. 3, 4 and 5 show that the temperature in the outer layers (Layer 1 and Layer 17) does not differ significantly. It can also be seen, that with higher heating temperatures the temperature recorded in the core of the part (Layer 9) increasingly deviates from the temperature in the outer layers (Layer 1 and Layer 17). Fig. 6 illustrates this more clearly.



Figure 6: Comparison of the core temperatures (Layer 9) at three different heating temperatures.

While the core temperature of the sample heated to 200°C reaches a plateau due to machine control as the outer layers reach the target temperature and the machine switches to reduced heat input, the slope of the core temperatures at 250°C and 300°C heating temperature flattens due to reduced heat transfer influenced by deconsolidation and air pockets.

To investigate the assumption of higher deconsolidation at higher heating temperatures, the part thickness was measured after the heating experiments and compared with the initial thickness before heating (see Table 1 and Fig. 7). The initial thickness is a result of the consolidation process and is assumed to be homogenous across the part, without significant deviations.

Heating temperature	Initial thickness	Mean thickness after heating	Thickness at sensor position	Difference
200°C	3.20 mm	3.87 mm	4.00 mm	0.67 mm
250°C	3.12 mm	3.98 mm	4.38 mm	0.86 mm
300°C	3.06 mm	4.00 mm	4.40 mm	0.94 mm

Table 1: Mean values of initial and final thickness, and the difference before and after heating.

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Figure 7: Overview of the thickness difference before and after heating – mean values and at position of the sensor placed in the core of the composite part.

Fig. 7 illustrates the increase in thickness with higher heating temperatures. The relatively high standard deviation leads to the assumption, that deconsolidation is a local phenomenon, and that the thickness of the preheated part varies significantly. It further shows that the position of the sensor placed at the core of the composite results in an even greater change in thickness during heating. This is because the sensor acts as a defect, causing internal stresses during the cooling step in consolidation. These stresses are released when the part is heated and result in deconsolidation. Therefore, the slopes of the 250°C and 300°C heating temperature curves in Fig. 6 show the significant change when heated above 200°C.

#### 4.1.2 Measurement of the thermal diffusivity

The results of the active thermography measurements are shown in Fig. 8, where the thermal diffusion time is illustrated over a reduced sample area ( $95x197mm^2$ ), caused by the fixation frame. It can be seen that the diffusion time is inhomogeneous across the part and certain areas with high diffusion times can be identified as deconsolidated spots.



Figure 8: Thermal diffusion time of parts after preheating at various temperatures: a) no preheating, b) preheating at 200°C, c) preheating at 250°C, and d) preheating at 300°C.

To obtain a representative value for each part, the mean value of the thermal diffusion time over the measured sample was used and the thermal diffusivity a was calculated for each part using Eq. (3).

Since the simulation is not able to account for thickness variation, i.e. deconsolidation, during heating, which affects heat transfer, the initial part thickness was used to calculate the average thermal diffusivity *a*. The results are listed in Table 2. Note that to measure the thermal diffusion time for a part heated to  $300^{\circ}$ C, a representative 2 mm thick sample was used. The reason for this was that the 3 mm thick preheated specimens were highly deformed due to the release of internal stresses during heating, which was a major complication in the measurement procedure. Therefore, the diffusion time for the sample preheated at  $300^{\circ}$ C is lower than that for the parts preheated at  $250^{\circ}$ C and  $200^{\circ}$ C, respectively, due to the reduced thickness.

Heating temperature	Mean value of the diffusion time	Standard deviation of the diffusion time	Mean value of the thermal diffusivity <i>a</i>
No preheating	11.48 s	0.74	3.49e-7 m <sup>2</sup> /s
200°C	46.29 s	7.39	1.94e-7 m <sup>2</sup> /s
250°C	57.88 s	9.19	1.55e-7 m <sup>2</sup> /s
300°C	30.01 s	12.57	1.33e-7 m <sup>2</sup> /s

Table 2: Calculated thermal diffusivities obtained by measurement of the diffusion time for the investigated parts.

#### 4.2 Modeling results

The simulation results show a very good agreement with the data obtained from the heating experiment up to 200°C (see Fig. 9 – 11). As mentioned above, the layup starts to deconsolidate to a large extent around 200°C, especially at the position of the sensors, as they act as an inclusion. Since the simulation is unable to model deconsolidation due to inclusions and the associated change in heat transfer, the temperature rise is largely unhindered up to the plateau of the target temperature.



Figure 9: Comparison of the temperature between experiment and simulation for a) layer 1 and b) layer 9 at a preheating temperature of 200°C.



Figure 10: Comparison of the temperature between experiment and simulation for a) layer 1 and b) layer 9 at a preheating temperature of 250°C.



Figure 11: Comparison of the temperature between experiment and simulation for a) layer 1 and b) layer 9 at a preheating temperature of 300°C.

It is assumed that the thermodynamic behavior of the composite part at an arbitrary position without an artificial inclusion, i.e. a temperature sensor, is similar to the behavior suggested by the simulation. However, to the best of the authors' knowledge, there is no alternative to recording the temperature at various positions within a composite layup, but with sensors that amplify deconsolidation.

## 9 CONCLUSIONS

In this work, a simulation which only uses the Laplace equation was used to determine the temperature at the core of a UD tape layup during preheating. To this end, samples of polycarbonate with 44% carbon fiber by volume were heated in an infra-red oven at 200°C, 250°C, and 300°C and their thermal diffusivity was determined by active thermography. To validate the simulation temperatures at the outer layers and the core were recorded during the heating experiments.

The experiments showed that at higher heating temperatures more deconsolidation occurs, which reduces the heat transfer from the part surface to the core. However, due to a relatively high standard deviation, it can be concluded, that deconsolidation is a local phenomenon and occurs inhomogeneously across the part. Furthermore, the investigations showed that the sensor at the core of the composite promotes further deconsolidation. The thickness change at the position of the sensor was significantly

higher than the mean values, which leads to invalid temperature values. It is assumed, that the reason for this high thickness changes was the release of internal stresses that occurred during the consolidation step, presumably caused by the sensor wire.

Due to the lack of reproducibility of local deconsolidation phenomena an average value of the thermal diffusivity of each part was determined and used for the simulation to obtain a reasonable prediction of the temperature rise during heating.

The results of the simulation showed that the temperature in the core of the part can be well predicted up to the point where extensive deconsolidation starts due to the sensor acting as an inclusion. It can be assumed that the temperature at a position without an inclusion behaves according to the simulation, but this needs to be proven.

For the future, further investigations on preconditioning and preprocessing of the composite parts and their influence on deconsolidation and heat transfer during heating are worthwhile.

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#### REFERENCES

- [1] A. Benatar, and T.G. Gutowski, "Methods for Fusion Bonding Thermoplastic Composites", SAMPE Quarterly. 18(1): 35–42 (1986).
- [2] S.M. Todd, "Joining Thermoplastic Composites", 22nd International SAMPE Technical Conference, pp. 383–392 (1990).
- [3] G. J. Sweeney, P. F. Monaghan, M. T. Brogan and S. F. Cassidy "Reduction of infra-red heating cycle time in processing of thermoplastic composites using computer modelling". Composites Manufacturing, Vol. 6, No. 3-4, pp 255-262, 1995.
- [4] R. Akkerman and S.P. Haanappel "6 Thermoplastic composites manufacturing by thermoforming". Advances in Composites Manufacturing and Process Design, Woodhead Publishing, pp 111-129, 2015.
- [5] M. Pettersson and S. Stenström "Modelling of an electric IR heater at transient and steady state conditions: Part I: model and validation". International Journal of Heat and Mass Transfer, Vol. 43, No. 7, pp 1209-1222, 2000.
- [6] G. Mayr, B. Plank, J. Sekelja and G. Hendorfer "Active thermography as a quantitative method for non-destructive evaluation of porous carbon fiber reinforced polymers". NDT & E International, Vol. 44, No. 7, pp. 537-543, 2011.
- [7] OpenCFD Ltd. "laplacianFoam", accessed 6.10.2022, https://www.openfoam.com/documentation/guides/latest/doc/guide-applications-solvers-basic-laplacianFoam.html.